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Submitted to:

http://lib-www.lanl.gov/la-pubs/00818457.pdf

USING A MULTI-DISCIPLINARY APPROACH, THE FIRST ELECTRON BACKSCATTERED KIKUCHI PATTERNS WERE CAPTURED FOR A PLUTONIUM ALLOY

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ABSTRACT

Microstructural characterization of plutonium by electron backscattered diffraction (EBSD) has been previously elusive primarily because of the extreme toxicity and surface oxidation rates associated with plutonium metal. In this work, initial electron backscattered diffraction pattern (EBSP) observations of a plutonium-gallium (Pu-Ga) alloy were made. Samples were prepared using standard metallographic and electropolishing techniques that were performed inside gloveboxes and/or an open front hood to prevent spread of radioactive contamination. A scanning Auger microprobe (SAM), equipped with an ion-gun, was used to characterize and remove surface chemical impurities (in particular carbon (C) and oxygen (O)) and a specially designed vacuum transfer device was used to minimize oxidation during the sample transfer from the SAM to the scanning electron microscope (SEM). EBSD patterns of the δ-phase (face-centered-cubic) were captured and the experimental techniques and parameters used to perform EBSD characterization are described in detail.

INTRODUCTION

The relative crystallographic orientations of separate grains within a material, also called crystallographic texture, have long been known to strongly influence material properties. The typical means of obtaining bulk texture data has been through the use of X-ray or neutron diffraction, while transmission electron microscopy (TEM) has provided local texture information. Recent technological advances in microscopy techniques have provided a means to evaluate crystallographic texture using common metallurgical preparation techniques. This development has been in the field of EBSD mapping through the automated collection and indexing of EBSPs [1]. This SEM technique is briefly described as follows. When incident electrons penetrate into a sample, they may experience a variety of elastic scattering events until the sample absorbs their energy or they may scatter elastically and escape from the specimen surface as backscattered electrons (BSEs). The diffracted BSEs are emitted in a series of bands, known as Kikuchi bands, which relate to the crystal planes and their orientation within the sample. For EBSP capture, a phosphor screen is placed in the SEM vacuum chamber close to the sample, which is steeply inclined to maximize the contrast in the BSEs used to produce the diffraction patterns. When the BSEs bombard the phosphor screen, a network of the Kikuchi bands is then imaged by a camera. The positions of the bands, which contain information relating to the symmetry and orientation of the crystal, are then identified. Sharp EBSPs are likely formed entirely from BSEs that have experienced only elastic interaction. Thus, EBSD is

a surface sensitive technique and quality of the patterns is strongly dependent on sample preparation. This is especially true for the high-density materials such as Pu. For example, from Monte Carlo-based simulations and inelastic mean-free path theory, it was estimated that 95% of the incident electrons would experience an inelastic collision (i.e. structural information lost) within 9nm of the surface for δ-Pu (ρ=15.92g/cm^3). Thus the bulk of the EBSD information for Pu is expected to come from only the top several nanometers of the surface.

Some of the advantages of this technique include the large number of structural orientation measurements attainable in a relatively short period of time, the ability to nondestructively characterize local areas of relatively large samples, and the relatively straightforward specimen preparation. The latter allows for the collection of a variety of data (EDS and WDS, X-ray and neutron diffraction, and SEM characterization to name just a few) for identical areas of the same sample. In addition, the equipment used for EBSD is easily adaptable to most SEMs and is relatively inexpensive[1]. The first commercially available system was introduced in 1994, and since then the growth of sales worldwide has been dramatic. This has been accompanied by a widening applicability to materials science problems such as microtexture, phase identification, grain boundary character distribution, deformation microstructures, and phase transformations [2].

EBSD analysis is ideally suited for Pu investigations because of the complex phases and phase transformations possible. In particular, EBSD would be useful in its ability to address some of the many questions concerning the microstructural evolution, aging, and phase transformation behavior in Pu and its alloys. However, EBSD analysis of Pu has been elusive to date due to several factors including rapid surface oxidation. This process results in the buildup of an amorphous surface layer that acts as an obstacle for electron penetration to the underlying crystalline metal. As described here, a multidisciplinary approach was used to overcome the obstacles preventing EBSD analysis of Pu.

METHODS

The material used in the initial evaluation was a δ-phase (face-center-cubic (fcc) structure) Pu-Ga alloy, see Figure 1. For most nonreactive metals, simple metallographic polishing followed by atmosphere transfer to the SEM is sufficient for EBSD capture. However, this is not the case for highly reactive metals such as Pu and additional surface cleaning, such as ion sputtering, is necessary. For this reason a multifunctional ultra-high vacuum SAM (see Figure 2) was used to characterize and remove surface layers contaminated with chemical impurities. Figure 3 shows the Auger spectra taken from the surface of the uncleaned sample. In addition to small Pu peaks, large C and O peaks and a small sulfur peak were observed. In order to remove the surface layers concentrated with contaminants, sequential treatments of argon (Ar) ions were used to bombard the sample surface. Comparing the Auger spectra of Figure 3, the elimination of the impurities to below detection limits (<0.1 atomic percent) and the increased height of the Pu peaks were evident for the sputtered condition.

Most environmentally-sensitive materials which require sputter cleaning can be transferred from the ion-cleaner through air to the SEM chamber. For materials which oxidize rapidly, this jeopardizes the surface integrity. For the Pu-Ga sample, atmospheric transfer did not result in successful EBSP acquisition, and this was attributed to the formation of a thick

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[1] For example, the cost for an SEM with EBSD capability pales in comparison to that of a TEM.

amorphous surface oxide layer as a result of air exposure. Even an atmospheric exposure of only one second after ion-sputtering led to a surface which did not exhibit EBSPs of the underlying metal. In order to minimize surface oxidation, a specially designed transfer device, including a vacuum suitcase and SEM entry-port adapter (see Figure 4a), was designed to transfer the sample to the SEM chamber (see Figure 4b) under vacuum.

RESULTS AND DISCUSSION

Figures 5a and b are SEM images representative of the ion-sputtered microstructure. Along with removing the surface layers concentrated with C and O, ion-sputtering enhanced the grain boundary contrast. The grains were topographically distinct, which is a result of the different sputtering rates as a function of crystallography. One of the most interesting aspects of the sputtered microstructure was that the bulk impurities tended to sputter at a different rate than the δ-phase metal. This resulted in tooth-like features which protruded up to 2μm above the surface (see Figure 5b). EDS surveys and microscopy suggested that the surface impurities shadowed the underlying Pu-Ga metal, resulting in the protrusions. SEM examination partway through the sputtering series revealed a growing number of the fine protrusions, in agreement with the idea that the impurities were not introduced during metallographic preparation and that ion-sputtering removes the δ-phase metal at a faster rate than the impurities. By minimizing sputtering to remove only those surface layers concentrated with C and O, the surface protrusions, which interfere with the BSEs used for EBSD, are minimized.

EBSPs were obtained from different δ grains, see Figure 6. After the initial patterns were captured, the sample was left in the SEM chamber for 68 hours and the sample was then reanalyzed successfully for EBSPs. This indicates that the surface oxide did not thicken enough within the vacuum environment to prevent EBSD observation of the Pu-Ga metal. Combining this observation with those for the air transfers, the effect of environment on surface oxidation is illustrated, and this indicates the importance of maintaining a low-oxygen environment after surface cleaning for EBSD observation of Pu-Ga alloys. Overall, this demonstrated sample preparation and characterization technique, involving several disciplines including metallography, microscopy, and surface science, is expected to provide a powerful means to further understand phase transformation behavior, orientation relationships, and texture in the complicated Pu and Pu-alloy systems.

CONCLUSIONS

(i)   The first EBSPs were captured for a Pu-Ga alloy.
(ii)   Sputtering the surface with Ar ions and transferring the cleaned sample to the SEM in a vacuum environment minimized surface oxidation. This maintained the surface integrity of the sample and allowed for the capture of EBSPs of the fcc δ phase.

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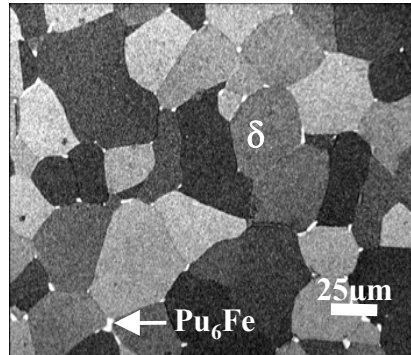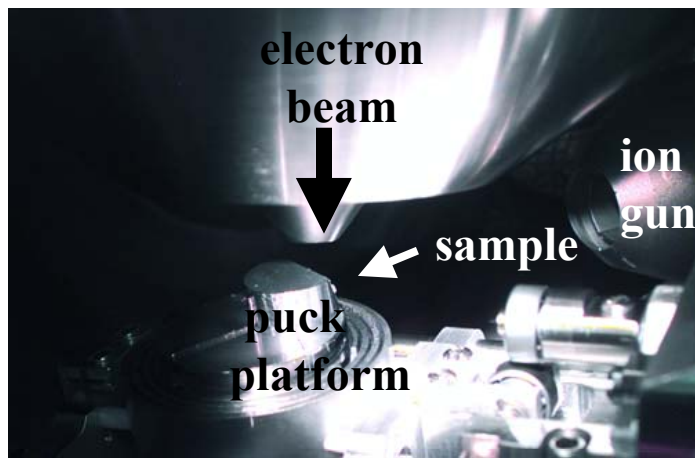
Figure 1. The Pu-Ga alloy microstructure.

Figure 2. A 3mm diameter sample (not seen) sits on a puck assembly inside the chamber of the ultra-high vacuum scanning Auger microprobe, which is capable of sputtering materials using a variety of ions, ion energies, and sputtering angles.
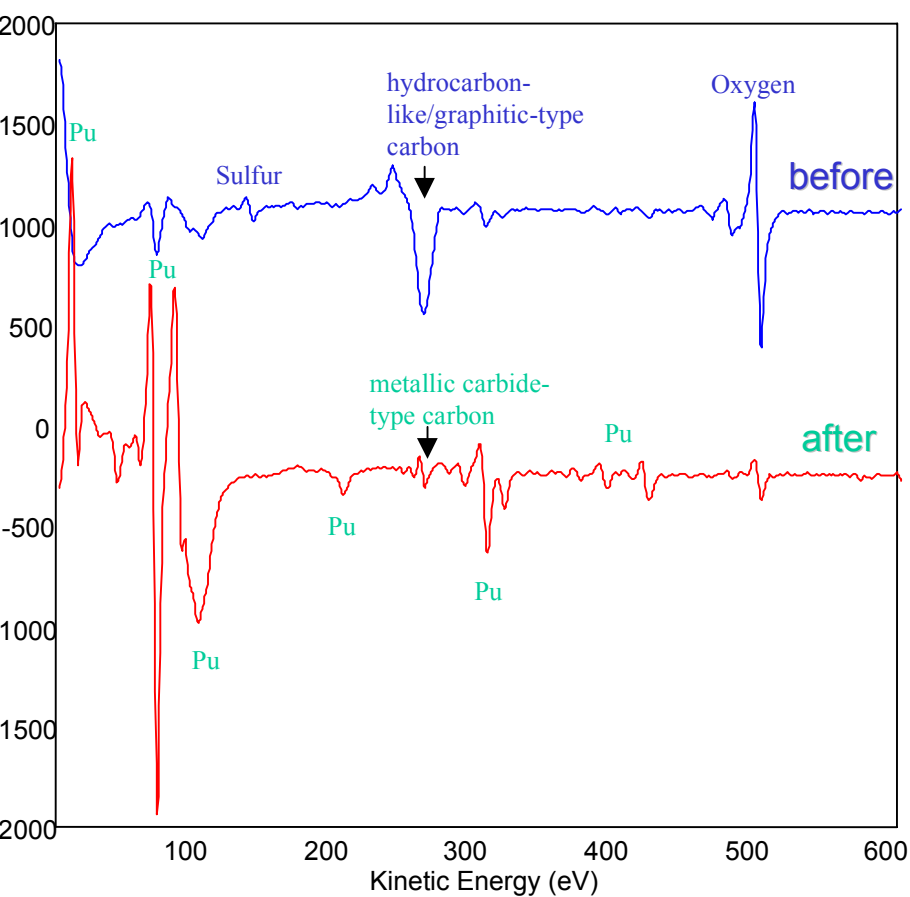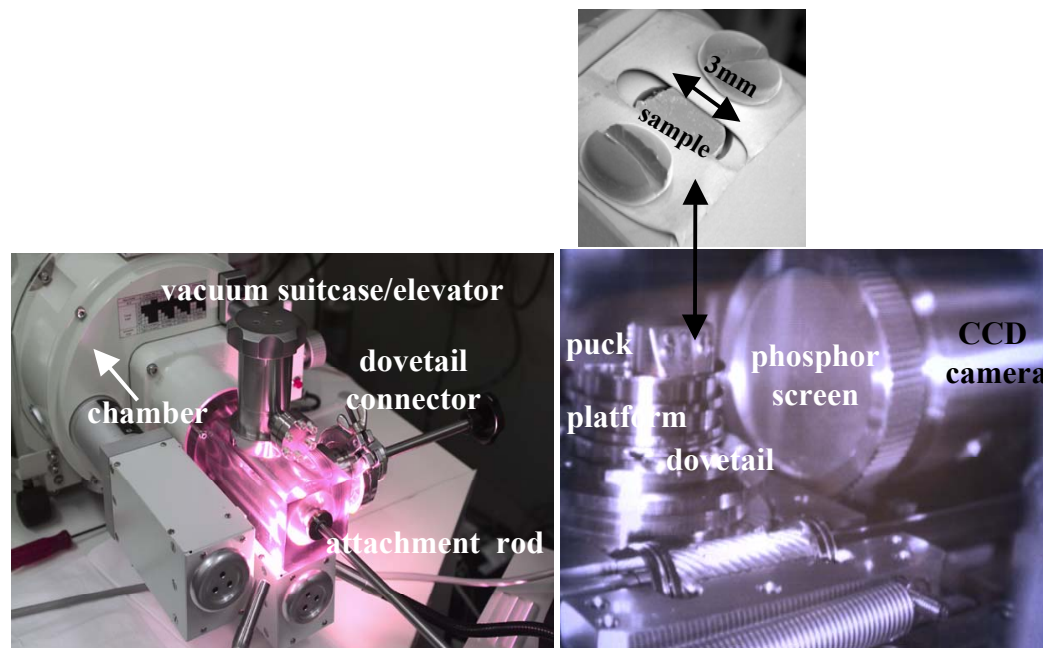
Figure 3. Auger spectra before and after Ar ion-sputtering the sample surface. The large C and O peaks observed before sputtering were nearly eliminated during the sputtering procedures.

(a) (b)

Figure 4. In order to minimize oxidation during sample transfer, a (a) vacuum suitcase and SEM entry-port adapter were used and the sample was transferred to the (b) SEM chamber in a vacuum.
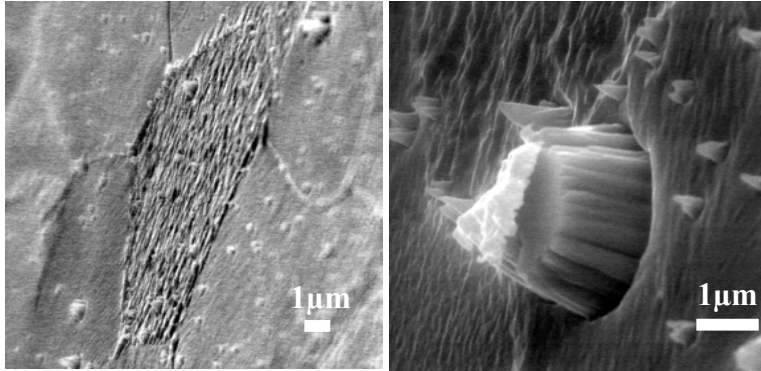
(a)   (b)

Figure 5. (a) Low- and (b) high-magnification SEM images of the ion-sputtered microstructure. The material inclusions sputtered at a different rate than the Pu-Ga metal and the surface roughness of the grains is a result of the effects of crystallographic orientation on ion-sputtering. Note that the sample normal was tilted 70° with respect to the beam.
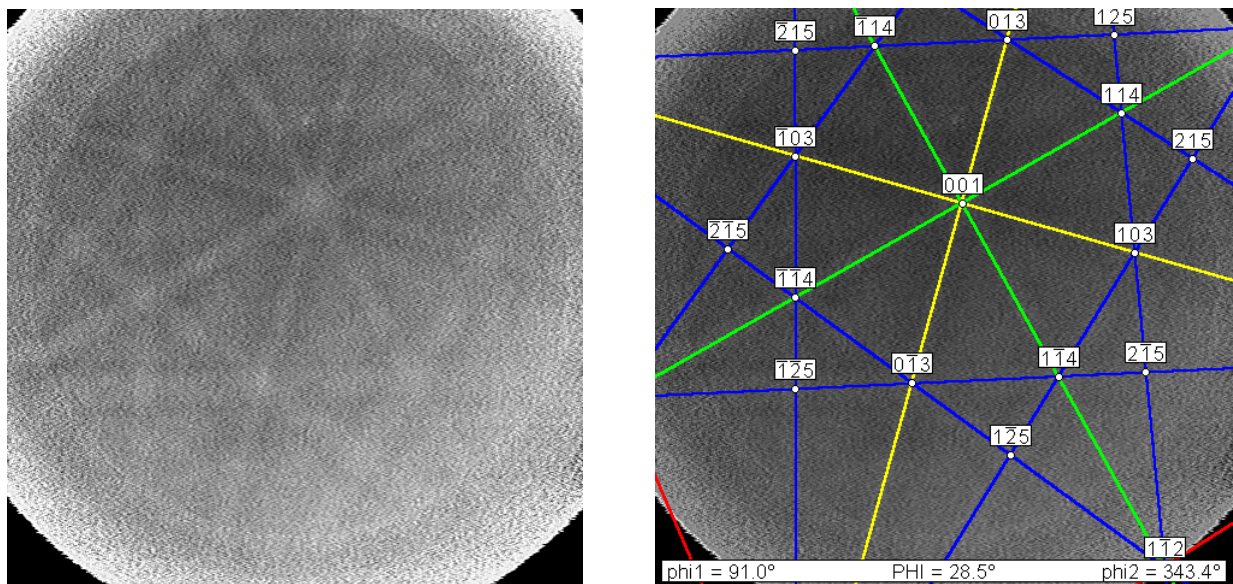
Figure 6. Electron backscattered Kikuchi pattern and its indexed orientation.